J. of the Korean Radioactive Waste Society
Vol.3(4), P. 329-333, Dec. 2005.

Effect of engineered barriers on the leach rate of cesium from spent PWR fuel

가압경수로 사용후핵연료 중 세슘의 침출에 미치는 공학적 방벽 영향

Kwan Sik Chun, Seung-Soo Kim and Jong-Won Choi
Korea Atomic Energy Research Institute, 150 Duckjin-dong, Yuseong-gu, Daejeon

kschun@kaeri.re.kr

전관식, 김승수, 최종원
한국원자력연구소, 대전시 유성구 덕진동 150번지

(Received June 21, 2005 / Approved September 1, 2005)

Abstract

To identify the effect of engineered barriers on the leach rate of cesium from spent PWR fuel under a synthetic granitic groundwater, the related leach tests with and without bentonite or metals have been performed up to about 6 years. The leach rates were decreased as a function of leaching time and then became a constant after a certain period. The period in a bare spent fuel was much longer than that with bentonite or metal sheets. The cumulative fraction of cesium released from the spent fuel with bentonite or with copper and stainless steel sheets was steadily increased, but the fraction from bare fuel was rapidly and then sluggishly increased. However, the values deducted its gap inventory from the cumulative fraction of cesium released from the bare fuel was almost very close to the others. These suggest that the initial release of cesium from bare fuel might be dependant on its gap inventory and the effect of engineered barriers on the long-term leach rate of cesium would be insignificant but the rate with engineered barriers could be reduced in the initial transient period due to their retardation effect. And the long-term leach rate of cesium from spent fuel in a repository would be approached to a constant rate of 2×10^{-2} g/㎡ -day.

Key words : leach rate, spent fuel, cesium, bentonite, metal sheets, repository

요 약

모의지하수중에서 가압경수로형 사용후핵연료로부터 세슘의 침출률에 미치는 공학적 방벽 영향을 규명하기 위하여 지난 약 6년간 벤토나이트 또는 금속시편등의 존재여부에 따라 침출시

험을 수행하였다. 침출률은 시간이 경과함에 따라 지수함수적으로 감소하는 경향을 나타내었으며, 적정시간이 경과한 다음부터는 일정한 값에 수렴하는 경향을 나타내었다. 벤토나이트 또는 금속시편이 있을 경우 세슘의 누적누출분률은 선형적으로 증가하였으나, 이들이 없는 경우의 누적분률은 급격히 증가한 다음 서서히 증가하는 경향을 보였다. 이 누적분률에서 갭에 존재하는 세슘의 재고량을 제한 값은 공학적 방벽이 존재하는 경우의 누적분률에 거의 근접하였다. 이러한 결과들은 사용후핵연료 중 세슘의 초기누출분률은 갭 중 세슘의 재고량에 의존하지만, 세슘의 장기침출률은 공학적 방벽에 거의 영향을 받지 않음을 암시해 주고 있다. 그러나 세슘의 초기침출률은 공학적 방벽의 지연효과로 감소될 수 있을 것이다. 그리고 처분장에서 사용후핵연료 중 세슘의 장기침출률은 2×10^{-2} g/㎡ -day를 넘지않는 범위 내에서 일정한 값을 가질 것이다.

중심단어 : 침출률, 사용후핵연료, 세슘, 벤토나이트, 금속시편, 처분장

Ⅰ. Introduction

Many experimental results have been reported for the dissolution behavior of spent fuel and unirradiated UO_2 in water under various conditions [1-6]. Most of the studies on the dissolution of unirradiated UO_2 pellet with bentonite have been carried out and only a few experiments have been concerned with the identification of dissolution mechanism of spent nuclear fuel under repository conditions [7-10], which is not well known with the lack of real data. In KAERI, the long-term leaching experiment of spent PWR fuel has been carried out in synthetic granitic groundwater since June of 1998. The purpose of this experiment is to get the information on the dissolution behavior of spent fuel and the long-term leach rate of radionuclides from the fuel under a repository condition in order to support the development of radionuclide release model from spent fuel in the near field and to identify the release mechanism of radionuclides, and finally to support the performance assessment of our reference disposal concept.

This paper covers the effect of waste package materials and bentonite block, which could be considered as an engineered barrier, on the leach rate of cesium from spent PWR fuel under synthetic granitic groundwater and also the long-term leach rate of cesium is estimated in this paper as well, based on the empirical formula derived from the leach rates experimentally measured.

Ⅱ. Experimental

1. Preparation of specimens

The rod of spent PWR fuel was cut to be about 3mm thickness with a diamond blade. The burn-ups of the specimens were presumed to be 39,089 and 37,805 MWD/MTU for J44-H08 and J-44-A03 discharged from Kori-2 Nuclear Power Plant, respectively, and their initial enrichment of U-235 was 3.4869wt.%. Then all the specimens were weighted and their dimensions measured.

2. Leach test

Domestic Ca-bentonite, which was compacted to 1.4Mg/㎥, was filled into the under-part of a leach cell (43mmΦ × 33mm) except space to hold a specimen, and one specimen was put in the space. Then, the under-part was bolted with the upper-part in which only the compacted bentonite was filled. The cell was put into 450ml of synthetic granitic groundwater (Table 1) as leachant in a bath (135mmΦ × 180mm). The stainless steel 316L was used for the fabrication of the cells and the baths. And stainless steel filters with the pore size of 10㎛ were put at the top and bottom sides of the cell in order to prevent the release of the compacted bentonite into the leachant by its swelling. The leachant was purged with Ar-gas for over 15 minutes in order to remove oxygen, and all baths were kept in an Ar-filled box at a little higher pressure than that in the hot cell. For the comparison of the effect of bentonite on the leaching of spent fuel, some specimens without bentonite were put into baths. And two bare spent fuel fragments were loaded into the baths with and without sheets of copper and stainless steel. All the loaded specimens have been emplaced in a hot cell for their leach test at an ambient hot cell temperature (see Fig. 1).

3. Analysis

About 10ml of the leachate from each bath was periodically sampled and the cesium activity of this leachate was measured by using γ-spectrometer (EG&G ORTEC ADCAM-100). The copper and stainless steel sheets from the cell for the specimen were taken out, settled into the 5M HNO_3 solution for one day, and then the activity of the solution has been measured. Some cells with the bentonite blocks were dismantled after a certain period of their leaching and then the blocks were dissolved by the addition of concentrated HF, HNO_3, HCl at around 80℃ and their activities of cesium in the solutions were measured.

Table 1. Composition of synthetic granitic groundwater

Element	K	Sr	Mg	Li	Ca	Zn	Mn
Concentration (㎎/L)	0.75	0.19	0.58	0.09	10.5	0.08	0.01
Element	HCO_3	Cl	SO_4	CO_3	Na	F	Fe
Concentration (㎎/L)	74.9	18.9	25.3	9.9	50.8	7.69	0.04

Ⅲ. Results and discussion

The leach rates of cesium from spent fuel specimens up to 2124 days according to various circumstances such as with metal sheets or bentonite block or without any barriers are illustrated in Figure 2. This figure shows that the rates of cesium from the specimens were rapidly decreased up to a certain periods and then

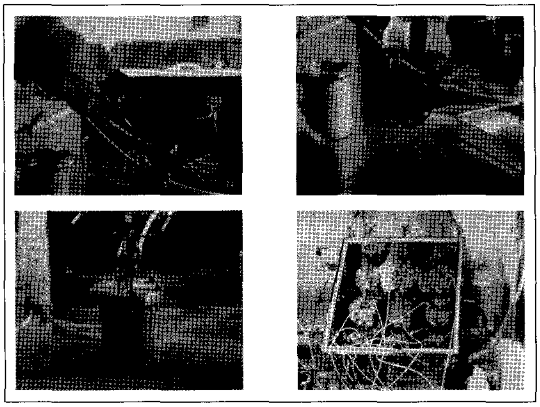

Figure 1. Photograph of leach cells and water baths in the hot cell

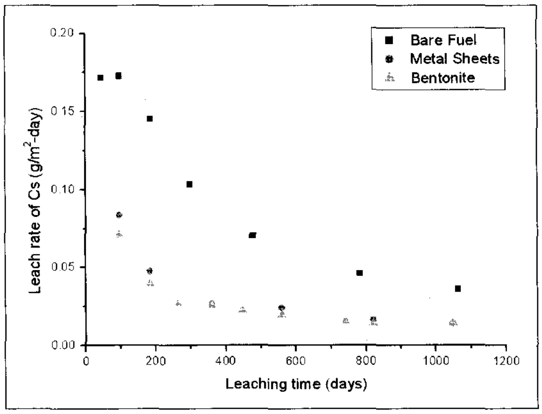

Figure 2. Leach rates of cesium as a function of leaching time

became a constant. Such results showed a very similar tendency to Forsyth's result[11]. In the presence of bentonite or metal sheets the constant rate would reach at around 600 days later, while in the bare spent fuel the time to reach a constant would probably require for over 1000 days. And at around 100 days the leach rates with bentonite and metal sheets were 8.3×10^{-2}g/m²-day and 7.1×10^{-2}g/m²-day, respectively, but the rate from the bare fuel (1.7×10^{-1}g/m²-day) was about two times higher than these values. This suggests that the initial leach rate could be retarded by the engineered barriers.

The cumulative fractions of cesium released from the specimens up to around 2124 days are shown in Figure 3. This figure shows that the fraction of cesium released from the bare fuel specimen was rapidly increased up to around 600 days and then sluggishly increased, while the fractions from the others were steadily increased and these fractions were very close each other. The fraction from the bare fuel up to about 600 days was about 7.7×10^{-3}, which was close to the gap inventory of cesium (6×10^{-3})[10,12], and up to about 2124 days is about 9.5×10^{-3}, which was higher than the results of Finn et al.[13,14] but lower than that of Forsyth[15]. The value deducted its gap inventory from the cumulative fraction of cesium released from the bare fuel was almost the same as the fractions from the others, 3.3×10^{-3} for only metal sheets and 3.4×10^{-3} for bentonite up to 2092 days. This suggests that the initial release of cesium from bare spent fuel would be dependent on its gap inventory, but the amount of the initial transient release could be reduced due to the presence of bentonite or waste package

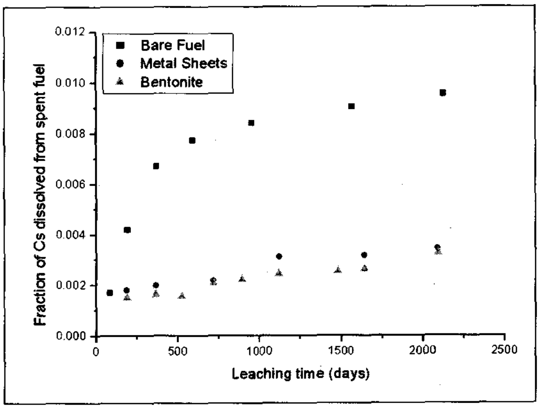

Fig. 3. Cumulative fractions of cesium released by leaching up to 2124days

materials. This may be due to the formation of alteration products that could incorporate cesium[16] or to the reduction of uranium solubility by oxygen scavenging effect of some materials.

From the measured values the empirical formulas of the leach rates of cesium are derived as the following :

For bare spent fuel :

$Ro = 1.81\times10^{-2} + 1.82\times10^{-1}Exp[-2.38\times10^{-3} * t]$ (1) and

For spent fuel in contact with bentonite blocks :

$Rb = 1.74\times10^{-2} + 1.40\times10^{-1}Exp[-8.09\times10^{-3} * t]$ (2), and

For spent fuel with metal sheets :

$Rm = 1.63\times10^{-2} + 1.21\times10^{-1}Exp[-8.57\times10^{-3} * t]$ ··· (3),

where Ro, Rb and Rm are the leach rates, as g/m²-day, and t is the leaching time, as days. These equations indicate that the long-term leach rate from the spent fuel specimen would be almost approached to around 2×10^{-2} g/m²-day. Even though these values were over three times higher than the range of Forsyth's results[11], our results suggest that the effect of engineered barriers on the long-term leach rate of cesium from spent PWR fuel would be insignificant, but the leach rates in the initial transient period could be retarded by these materials.

IV. Conclusions

To identify the effect of engineered barriers on the leach rate of cesium from spent PWR fuel under a repository condition, the related leach tests have been performed up to about 6 years.

The experimental results indicate that the effect of copper, stainless steel and bentonite on the long-term leach rate of cesium from spent fuel in a repository would be insignificant and that the initial transient leach rate could be retarded by engineered barriers. And the results also suggest that the initial release of cesium from a bare fuel might be dependent on its gap inventory.

The empirical formulas suggest that the long-term leach rate of cesium from spent PWR fuel in a repository would be below 2×10^{-2} g/ m²-day.

Reference

[1]J.A. Serrano et al., "Comparison of the leaching behavior of irradiated fuel, SIMFUEL, and Non-irradiated UO_2 under oxic conditions" , Radiochimica Acta, vol.82, p.33 (1998)

[2]K. Lelous et al., "Leaching of spent fuel and simulated fuel in the presence of environmental materials : Integral experiments", Mat. Res. Soc. Symp. Proc., vol.506, p.191 (1998)

[3]D.J. Wronkiewicz et al., "Ten-year results from unsaturated drip tests with UO_2 at 90℃ : Implications for the corrosion of spent fuel", J. Nucl. Mat., vol.238, p.78 (1996)

[4]R.S. Forsyth, L.O. Werne and J. Bruno, "Preliminary study of spent UO_2 fuel corrosion in the presence of bentonite", J. Nucl. Mat., vol.160, p.218 (1988)

[5]J.C. Tait, S. Stroes-Gascoyne, R.J. Porth,, D. Wood and R.B. Heimann, "Leaching of used CANDU fuel in the presence of clay, titanium, and granite rock", Mat. Res. Soc. Symp. Proc., vol.127, p.309 (1989)

[6] L.H. Johnson and J.C. Tait, " Release of segregated nuclides from spent fuel", SKB-TR-97-18 (1997)

[7]Y. Albinsson, R. Forsyth, G. Skarnemark, M. Skalberg, B. Torstenfelt and L. Werme, , (Mat. Res. Soc. Symp. Proc. 176, Boston, U.S.A., 1989) pp.559-565.

[8]H. Rameback, M. Skalberg, U.B. Eklund, L. Kjellberg and L. Werme, Radiochimica Acta 82, 167 (1998).

[9]K. Lelous et al., "Leaching of spent fuel and simulated fuel in the presence of environmental materials : Integral experiments", Mat. Res. Soc. Symp. Proc., vol.506, p.191 (1998).

[10]L.O. Werne et al., "Spent fuel performance under repository conditions: A model for use in SR-Can", SKB-TR-04-19 (2004).

[11] R. Forsyth, An evaluation of results from the experimental programme performed in the Studvik hot cell laboratory, 1997.

[12]S.S. Kim, K.S. Chun, Y.B. Kim and J.W. Choi, "Initial release of nuclides from spent PWR fuels", Proceedings of the 4th Korea-China joint workshop on nuclear waste management, Feb. 17-19, p238 (2004).

[13]P.A. Finn, R. Finch, E. Buck and J. Bates, "Corrosion mechanisms of spent fuel under oxidizing conditions", Materials Research Society Symposium proceedings, Vol.506, p.123 (1998)

[14]S.A. Steward, CSNF waste form degradation ; Summary absraction, ANL-EBS-MD-000015 Rev.00 (2000).

[15]R.S. Forsyth, "Spent fuel corrosion and dissolution", J. Nucl. Mat., vol.190, p.3 (1992).

[16]E.C. Buck et al., "A new uranyl oxide hydrate phase derived from spent fuel alteration", J. Nucl. Mater., vol.249, p.70 (1997).